

# WORLD INTELLECTUAL PROPERTY ORGANIZATION International Bureau



# INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6: C08K 5/523	A1	(11) International Publication Number: WO 96/11977  (43) International Publication Date: 25 April 1996 (25.04.96)
	US95/128	DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).
(22) International Filing Date: 12 October 199  (30) Priority Data: 13 October 1994 (13.10.9)		Published  With international search report.  US
(71) Applicant: AKZO NOBEL N.V. [NL/NL]; Velperv Box 9300, NL-6800 SB Arnhem (NL).	veg 76, P.	.o.
(71)(72) Applicants and Inventors: MOY, Paul, Y. [ Sandi Lane, Fishkill, NY 12524 (US). BRIGH A. [US/US]; 21 Zabela Drive, New City, NY 19	T, Daniel	lie,
(74) Agent: FENNELLY, Richard, P.; Akzo Nobel Inc. stone Avenue, Dobbs Ferry, NY 10522 (US).	., 7 Livin	ng-
·		
(54) Title: POLYCARBONATE-CONTAINING POLY	MERS F	LAME RETARDED WITH OLIGOMERIC PHOSPHATE ESTERS

#### (57) Abstract

Polycarbonate-containing compositions are flame retarded with a mixture which comprises an arylene-bridged oligomeric polyphosphate ester and an effective amount of an alkylene-bridged diphosphate compound for increased flame retardancy efficacy and improved processing characteristics.

# FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT AU BB BE BF BG BJ BC CF CG CH CI CM CS CZ DE DK ES FI FR GA	Austria Australia Barbados Belgium Burkina Faso Bulgaria Benin Brazil Belarus Canada Central African Republic Congo Switzerland Côte d'Ivoire Cameroon China Czechoslovakia Czech Republic Germany Denmark Spain Finland Prance Gabon	GB GE GN GR HU IE IT JP KE KG KP LI LK LU LV MC MD MG ML MN	United Kingdom Georgia Guinea Greece Hungary Ireland Italy Japan Kenya Kyrgystan Democratic People's Republic of Korea Republic of Korea Kazakhstan Liechtenstein Sri Lanka Luxembourg Latvia Monaco Republic of Moldova Madagascar Mali Mongolia	MR MW NE NL NO NZ PL PT RO RU SE SI SK SN TD TG TJ TT UA US UZ VN	Mauritania Malawi Niger Netherlands Norway New Zealand Poland Portugal Romania Russian Federation Sudan Sweden Slovenia Slovekia Senegal Chad Togo Tajikistan Trinidad and Tobago Ukraine United States of America Uzbekistan Viet Nam
--	---	---	---	---	--

1

# POLYCARBONATE-CONTAINING POLYMERS FLAME RETARDED WITH OLIGOMERIC PHOSPHATE ESTERS

### Background of the Invention

U.S. Patent No. 5,204,394 to J.C. Gosens et al. advocates the use of arylene-bridged oligomeric polyphosphate esters in the flame retarding of polycarbonate-containing polymer compositions. An oligomeric phosphate or blend of oligomeric phosphates are disclosed by the Gosens et al. patent as having the formula

where  $R_1$ ,  $R_2$ ,  $R_3$ , and  $R_4$  each represent an aryl or an alkaryl group chosen independently of each other and wherein X is an arylene group, m1, m2, m3, and m4, each independently of each other are 0 or 1 and wherein n=1, 2, 3, 4, or 5 or wherein, in the case of a blend of phosphates, n has an average value between 1 and 5.

#### Summary of the Invention

35

30

5

10

15

20

25

It has now been found that a mixture of an alkylene-bridged diphosphate compound and the foregoing type of arylene-bridged oligomeric polyphosphate ester is an effective flame retardant agent in the aforementioned types of polycarbonate-containing polymer compositions. It has been found that there is increased flame retardancy efficacy and improved processing characteristics for the mixture in such polymers.

5

10

15

20

25

30

2

#### Description of Preferred Embodiments

The polycarbonate-containing resins and the arylene-bridged oligomeric phosphate esters to which the present invention is applicable are described in the aforementioned U.S. Patent No. 5, 204,394 which is incorporated herein by reference in regard to such teaching.

The terminology "alkylene-bridged diphosphate compound" as used herein is to be understood to encompass monomeric and low oligomeric species of the formula

Where R is aryl, such as unsubstituted phenyl, n is a number ranging from 0 to about 5, and  $R^1$  is alkylene of from 1 to 8 carbon atoms.

invention contemplates using present comprising the arylene-bridged phosphate compositions and the alkylene-bridged compounds in a ratio of from about 1:2 to about polycarbonate-containing in the 2:1 weight basis on Generally speaking, the amount of such mixture compositions. used in the polycarbonate-containing compositions will vary from about 1 % to about 25 % on the weight of the polycarbonatecontaining composition.

The present invention is illustrated by the Examples which follow.

Effective Hea:

# EXAMPLES 1-5

These Examples set forth cone calorimetric data  $(35.0 \text{ kW/m}^2\text{ irradiance})$  on neat PC/ABS, neat resorcinol diphosphate (RDP) and blends of RDP and neopentylglycol diphosphate at various weight amounts.

The polymers composites were preblended to a five to one (PC/ABS) ratio and extruded in a twin screw extruder to a homogeneous blend. The phosphate flame retardant mixtures ("FR" in the Table given below) were preblended in the prescribed ratios before addition to the composites. The incorporation of the flame retardants was accomplished by metering the liquid components into the melt stream of the extruder through a side port injector. All flame retardant composites are formulated to contain a small amount of powdered TEFLON fluoropolymer antidrip agent.

TABLE 1

Dook West

	$\sim$	
,		
٠	v	

5

0

.5

			Release	Total Heat	of Combustic:
25	FR USED	%P	(kW/m²)	(KJ)	(MJ/kg)
	NONE	-	201	32.12	3.48
	RDP (Neat)	0.91	69	17.79	2.04
	RDP/NPGDP (2/1)	0.92	62	18.83	2.02
30	RDP/NPGDP (1/1)	0.71	52	14.66	1.61
	RDP/NPGDP (1/2)	0.80	43	16.04	1.85

WO 96/11977 PCT/US95/12801

4

### EXAMPLES 6-10

These Examples show the melt flow viscosity data (ASTM-1238, Condition "I", 230°C, 3.8 kg) for the blends tested in each of Examples 1-5, above.

	FR Sample (from Ex. 1-5)	g/10 minutes
	NONE	13.0
10	RDP (Neat)	16.9
	RDP/NPGDP (2/1)	18.8
	RDP/NPGDP (1/1)	15.3
	RDP/NPGDP (1/2)	17.3

5

The foregoing data is presented for purposes of illustrating certain embodiments of the present invention and, for that reason, should not be construed in a limiting sense. The scope of protection sought is set forth in the claims which follow.

# We Glaim: does not see the way to prove

5

10

15

20

25

- 1: A flame retarded polycarbonate-containing composition which comprises a mixture comprising (1) an arylene-bridged oligomeric polyphosphate ester and (2) an effective amount of an alkylene-bridged diphosphate compound for increased flame retardancy efficacy and improved processing characteristics.
- 2. A composition as claimed in Claim 1 wherein the alkylene-bridged diphosphate compound is of the formula

kata 15 m. Maria ng mga kata 112. An angwasi i

wherein R is aryl, n is a number ranging from 0 to about 5, and  $R^1$  is alkylene of from 1 to 8 carbon atoms.

3. A composition as claimed in Claim 1 wherein the alkylene-bridged diphosphate compound is of the formula

- where R is unsubstituted phenyl, n is a number ranging from 0 to about 5, and R<sup>1</sup> is alkylene of from 1 to 8 carbon atoms and the arylene-bridged ester and the alkylene-bridged compound are in a ratio of from about 1:2 to about 2:1 on a weight basis.
- 4. A composition as claimed in Claim 1 wherein the amount of such mixture used in the polycarbonate-containing compositions will vary from about 1 % to about 25 % on the weight of the polycarbonate-containing composition.

5. A composition as claimed in Claim 2 wherein the amount of such mixture used in the polycarbonate-containing compositions will vary from about 1 % to about 25 % on the weight of the polycarbonate-containing composition.

5

6. A composition as claimed in Claim 3 wherein the amount of such mixture used in the polycarbonate-containing compositions will vary from about 1 % to about 25 % on the weight of the polycarbonate-containing composition.

10

7. A composition as claimed in Claim 1 wherein the arylene-bridged oligomeric polyphosphate ester is resorcinol diphosphate and the alkylene-bridged diphosphate compound is neopentylglycol diphosphate.

15

8. A composition as claimed in Claim 3 wherein the arylene-bridged oligomeric polyphosphate ester is resorcinol diphosphate and the alkylene-bridged diphosphate compound is neopentylglycol diphosphate.

20

9. A composition as claimed in Claim 5 wherein the arylene-bridged oligomeric polyphosphate ester is resorcinol diphosphate and the alkylene-bridged diphosphate compound is neopentylglycol diphosphate.

25



### INTERNATIONAL SEARCH REPORT

International application No. PCT/US95/12801

A. CLASSIFICATION OF SUBJECT MATTER  IPC(6) :C08K 5/523 US CL :524/127			
According to International Patent Classification (IPC) or to both national classification and IPC			
B. FIELDS SEARCHED			
Minimum documentation searched (classification system for	ollowed by classification symbols)		
U.S. : 524/127			
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched			
	`		
Electronic data base consulted during the international sear	rch (name of data base and, where practicable, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVA	NT		
Category* Citation of document, with indication, wh	ere appropriate, of the relevant passages Relevant to claim No.		
Y US, A, 5,204,394 (GOSENS column 2, lines 5-41.	US, A, 5,204,394 (GOSENS ET AL.) 20 April 1993, see 1-9 column 2, lines 5-41.		
	US, A, 3,869,526 (COMBEY ET AL.) 04 March 1975, see 1-9 column 1, lines 1-29 and column 3, lines 29-50.		
JP, A, 59-202,240 (DAIHACHI KAGAKU KOG KK) 16 1-9 November 1984, see the abstract			
JP, A, 59-45,351 (ADEKA-ARGUS CHEM KK) 14 March 1-9 1984, see the abstract.			
*			
Further documents are listed in the continuation of E			
Special categories of cited documents:	"I" tater document published after the international filing date or priority date and not in conflict with the application but cried to understand the		
*A* document defining the general state of the art which is not considered principle or theory underlying the invention to be of particular relevance:  *X* document of particular relevance; the claimed invention cannot be			
*E* earlier document published on or after the international filing da	considered novel or cannot be considered to involve an inventive step		
cited to establish the publication date of another citation or other			
special reason (as specified)  document of particular relevance, the chained sevenance cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art			
P* document published prior to the international filing date but later than "&" document member of the same patent family the priority date claimed			
Date of the actual completion of the international search Date of mailing of the international search report			
31 JANUARY 1996	01 FEB 1996		
Name and mailing address of the ISA/US  Commissioner of Patents and Trademarks  Box PCT	Authorized officer		
Washington, D.C. 20231	VERONICA HOKE MAV		
English No. (703) 305-3230	1 1 # 1 # 1 M		

THIS PAGE BLANK (USPTO)